

Annihilation of point defects in nematic liquid crystals

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Point defects are likely to be generated in the process of filling a capillary tube subject to homeotropic conditions on the boundary. There is plenty of experimental evidence to hold that, when two defects with opposite topological charge happen to be closer than a critical distance, they attract each other. At first, they move very slowly; then, as the distance between them becomes less than a diameter, their relative speed increases dramatically, until they annihilate one other. In this paper we describe, by means of a simple dynamical model, the attraction and annihilation of two defects in an infinite tube. We write the balance between the rate of change in the elastic free energy and the energy dissipated in the director motion. Hence, we derive and solve the differential equation that describes the evolution in time of the distance between the defects. The outcomes of our analysis confirm many qualitative aspects of the experimental evidence. [S1063-651X(96)11111-9]

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I. INTRODUCTION

A *topological charge* may be assigned to all point defects in ordered media, as illustrated, for example, in [1,2]. Here, to be specific, we focus attention on nematic liquid crystals, though the main ideas underlying this paper could also be applied to other similar media.

When the manifold employed to describe nematics is the unit sphere S^2 , the topological charge of a point defect is a relative integer. Inspired by the suggestive analogy with electric charges, one expects that defects with opposite topological charge feel an attractive force, which draws one closer to the other, until they coalesce, and possibly annihilate each other, leaving no trace of their existence, if the total charge vanishes.

Sound mathematical reasons have been provided to show that equilibrium configurations with prescribed point defects of opposite charge cannot be stable, and must evolve in time reducing the distance between the defects, so as to reduce the elastic free energy stored in the region where the interaction between them takes place. Here we take the elastic free energy per unit volume as given by

$$\sigma_F = \frac{1}{2} K |\nabla \mathbf{n}|^2, \quad (1)$$

where K is a positive modulus and \mathbf{n} is the unit vector field representing the director; this is the form taken by the well-known Frank's formula in the *one-constant approximation*. Brezis, Coron, and Lieb show in [3] that the minimum of the elastic free energy in the whole space with prescribed point defects of a given charge is *not* attained, but there is a minimizing sequence with energy converging to the infimum of the energy functional, which for a pair of defects with charges $+1$ and -1 is proportional to the distance between them. Translated into the language of physics, this result means that the defects in a *topological dipole* are not at equilibrium (as the energy fails to attain its minimum), and the

force they feel is constant (as the infimum of the energy is proportional to the distance between them). A naïve argument to this effect was already brought up by Brinkam and Cladis in their review article [4]. To put this conclusion in the right perspective, the reader should heed that for them the director field around a topological dipole is subject to no boundary condition whatsoever, neither on the walls of a container, nor at infinity, as an asymptotic data. In the problem that concerns us below the dipole is within a capillary tube subject to homeotropic conditions for the director on the lateral boundary. Thus it should be no surprise if we find that the force experienced by the defects fails to be constant.

More generally, it was proven by Hardt, Kinderlehrer, and Lin in [5] that if there is a sequence of equilibrium configurations for the director, each involving point defects which come closer to each other as the sequence index grows, and eventually cancel out as the index diverges, then the defects cannot be stable for every configuration in the sequence. This simply means that the coalescence of defects, even when they move extremely slowly, cannot be regarded as a quasistatic process, being a sequence of equilibrium states unfit to describe it mathematically. Once more this teaches us the same lesson: coalescence and annihilation of defects is an intrinsically dynamical phenomenon.

In [6] Ericksen makes this point even clearer, calling upon an unpublished work by Cladis and Walters, who observed the evolution in time of a pair of opposite charged defects in a capillary tube so treated as to make the director normal to the wall: "Starting a bit less than 4 diameters apart, the two [defects] start moving toward each other, at a rate of a diameter per thousand minutes, slow enough to make it somewhat reasonable to consider using static theory to analyze the behavior at early times. However, when the distance between them becomes a bit less than a diameter, the relative velocity starts to increase dramatically, until they come together and annihilate each other."

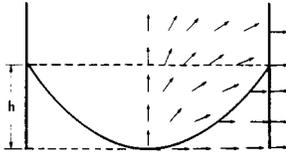


FIG. 1. Prototype of a *joint*, that is, an axisymmetric free surface which separates a peripheral region, where the director is a planar field, and a central region, where the director is an escaped field.

Our paper aims at proposing a mathematical model which describes this phenomenon through a peculiar dynamical system where the defects behave, in a sense, like particles. In Sec. II we recall from [7] and [8] a class of director fields in a capillary tube fit to mimic both a $+1$ defect and a -1 defect. The fields in this class are determined by the shape of a surface within the capillary, which we call the *joint*, where an orientation *escaped* along the axis and one pointing towards the lateral boundary are smoothly joined together. In Sec. III we determine the optimal joint for a point defect of either sign: it minimizes the elastic free energy stored in a cylinder sufficiently high to be treated as infinite. Though nonlinear, the equilibrium equation for the joint can be solved exactly, which makes our model ductile enough and its outcomes fully predictive. In Sec. IV we apply this model to a topological dipole. We minimize the elastic free energy when the distance between the defects is held fixed: within our special class the infimum of this functional is indeed attained; from it, we derive the force felt by the defects, which is proportional to the logarithm of the distance between them. In Sec. V, employing the dissipation principle posited in [9] by Leslie to give yet another derivation of the dynamical equations for nematics, we arrive at the differential equation that governs the relative motion of the defects, whence, in Sec. VI, we describe how they annihilate one another. In Section VII we collect the main conclusions of this paper, also attempting to compare the predictions of our mathematical model and the first experimental findings we are aware of: we discuss the issues now being addressed to make the agreement between them more than merely qualitative.

II. JOINTS

Here we describe a class of director fields exhibiting a point defect along the axis of a cylinder: we first focus attention on a $+1$ defect, a -1 defect being easily obtained from it. The fields we consider are all axisymmetric and subject to the homeotropic anchoring condition on the lateral boundary of the cylinder. The idea underlying the geometric construction employed below is finding an explicit junction between a field escaped along the axis, as the one discovered by Cladis and Kléman [10] and Meyer [11], and the radial field pointing towards the wall of the cylinder. This is achieved by introducing a *joint*, an axisymmetric free surface where the two fields adapt one to another without jumps. Figure 1 illustrates the prototype of such a joint: it separates two regions, one peripheral and the other central: the director is a planar radial field in the peripheral region, and an escaped field in the central region, differently rescaled on every section through a plane orthogonal to the axis. Clearly, at the

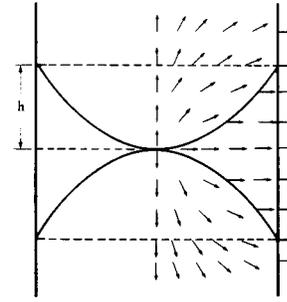


FIG. 2. Prototype of a $+1$ defect, composed by the joint in Fig. 1 and that obtained from it through a reflection.

lowest point of the joint there is a defect of the director, as it is a point where the director should simultaneously be parallel to the axis and to all radii.

Reversing upside down both the joint and the arrows representing the director field in Fig. 1, we obtain the prototype of a $+1$ defect, as illustrated in Fig. 2. In the class we employ, the two joints converging on the defect need not be symmetric, nor need they meet smoothly, as implied in Fig. 2. The shape of each joint is completely free and will be determined below by solving a variational problem: it may or may not bear an angular point on the defect. In Fig. 2 we see a $+1$ defect; a -1 defect is obtained from it by reversing all the arrows upside down relative to the transverse section of the cylinder, so that a domain escaped upwards turns into one escaped downwards, and vice versa. It is thus clear how a sequence of alternating defects can be generated by using fields alternately reversed.

We now give an explicit representation for the building block of this construction. We first consider a cylindrical tile, which will then be repeatedly employed along a capillary tube: we denote by R its radius and by h its height. We put the defect at the center of its lower base, as in Fig. 1, and we represent in cylindrical coordinates the longitudinal section of a joint through the function $r=r_0(z)$, where z varies in the interval $[0,h]$ while r_0 takes values in the interval $[0,R]$.

In the local frame of cylindrical coordinates $(\mathbf{e}_r, \mathbf{e}_\vartheta, \mathbf{e}_z)$ we express the director \mathbf{n} through the formula

$$\mathbf{n} = \cos\varphi \mathbf{e}_r + \sin\varphi \mathbf{e}_z, \quad (2)$$

where the angle φ is given by

$$\varphi(r, \vartheta, z) = \begin{cases} \frac{\pi}{2} - 2 \arctan\left(\frac{r}{r_0(z)}\right) & \text{for } 0 < r < r_0(z) \\ 0 & \text{for } r_0(z) \leq r \leq R. \end{cases} \quad (3)$$

In the inner region delimited by the joint, Eq. (3) (top line) represents the well-known field of Cladis and Kléman in a cylinder of radius $r_0(z)$. If $r_0(h)=R$, as in both Fig. 1 and Fig. 2, the field on top of the joint is the usual Cladis and Kléman's, which can be smoothly extended in a cylinder of radius R as a field independent of z , radially oriented only on the lateral boundary. It is indeed the presence of the joint that makes φ depend on z .

By (3) the elastic free energy stored in both regions adjacent to the joint ultimately depends on the function r_0 ,

which describes the shape of the joint. We denote by \mathcal{E}_h this energy, which is the integral of (1) over the cylinder of radius R and height h . As shown in Sec. 2 of [8], where the mathematical details of this class of director fields have already been presented, $\mathcal{E}_h = \pi K E_h$, where E_h is a functional of r_0 :

$$E_h[r_0] := \int_0^h \left\{ \alpha(r_0')^2 - \ln\left(\frac{r_0}{R}\right) + 2 \right\} dz \quad (4)$$

with a prime denoting differentiation with respect to z and $\alpha := 2 \ln 2 - 1$ a positive number magically emerging from the integration of the energy density over the radial variable. We often call E_h the energy of a joint of height h : it is invariant under reflection of both the joint and the director.

The Euler-Lagrange equation for E_h can easily be integrated once, leading to

$$\alpha(r_0')^2 = -\ln\left(\frac{r_0}{R}\right) + C, \quad (5)$$

where C is an arbitrary constant. The condition this equation is subject to for $z=0$ is clearly

$$r_0(0) = 0, \quad (6)$$

which prescribes the point where the defect lies, whereas that for $z=h$ requires a few words to be justified. It is

$$r_0'(h) = 0, \quad (7)$$

which represents the natural equilibrium condition at a free end point, as the radius $r_0(h)$ of the cross section of the joint where it reaches the prescribed height h is free to be so chosen as to minimize the energy E_h . Let

$$\rho := \frac{r_0(h)}{R}. \quad (8)$$

It follows from (5) and (8) that

$$C = \ln \rho, \quad (9)$$

which makes ρ the constant to be determined in $[0,1]$. By separation of variables, Eq. (5) can be solved for the inverse of the function $r_0(z)$; denoting such a function by $z_0(r)$, we obtain

$$z_0(r) = \rho R \sqrt{\alpha} \int_{\ln(\rho R/r)}^{+\infty} \frac{e^{-x}}{\sqrt{x}} dx, \quad (10)$$

which is our explicit representation for an equilibrium joint. It satisfies (6); moreover, by (8), it follows from (10) that

$$h = \rho R \sqrt{\alpha} \int_0^{+\infty} \frac{e^{-t}}{\sqrt{t}} dt = \rho R \sqrt{\alpha \pi}. \quad (11)$$

Thus, ρ is given a unique value, which through (10) determines a unique joint, provided that $h \leq \sqrt{\alpha \pi} R$. The shape of this joint is smooth everywhere. In particular, it is flat on the defect and tangent to the cylinder of radius $(h/\sqrt{\alpha \pi})$, where

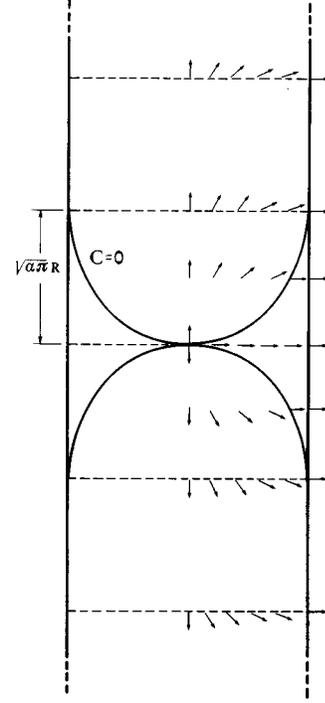


FIG. 3. A +1 defect structure in a cylinder of height $H = 4\sqrt{\alpha \pi} R$. The joints reach their maximum height and end tangent to the cylinder.

it reaches the height h , as is easily seen by taking the limits for $r \rightarrow 0$ and $r \rightarrow \rho R$ in the derivative with respect to r of the function in (10).

We compute the energy of the equilibrium joint by using (5) in (4), where the integration variable is changed into z with the aid of (10); recalling (9) also, we arrive at

$$E_h = (3 - \ln \rho) \rho R \sqrt{\alpha \pi}, \quad (12)$$

where ρ is related to h through (11).

If $h > \sqrt{\alpha \pi} R$, there is no equilibrium joint of height h . In our model, this lack of existence, typical of nonlinear variational problems, will have an effect on the interaction between two opposite charged defects. As $\sqrt{\alpha \pi} \approx 1.101$, the defects feel no force when they are farther apart than approximately 1.1 diameter of the capillary. Before drawing the dynamical consequences of this fact, in the following two sections we apply our construction to two simple static situations.

III. SINGLE DEFECT

Consider an infinite cylinder of radius R , which accommodates a +1 defect along its axis and enforces the homeotropic anchoring for the director on the lateral boundary. Figure 3 illustrates a portion of this cylinder, whose height is taken to be $H = 4\sqrt{\alpha \pi} R$ for reasons which will become clear below. Being higher than $2\sqrt{\alpha \pi} R$, there is no pair of equilibrium joints stretching through the whole of it on both sides of the defect. The highest equilibrium joint corresponds to choosing $\rho = 1$ (that is, $C = 0$) in (10); by (11), its height is $\sqrt{\alpha \pi} R$. Both joints in Fig. 3 are thus tangent to the cylinder;

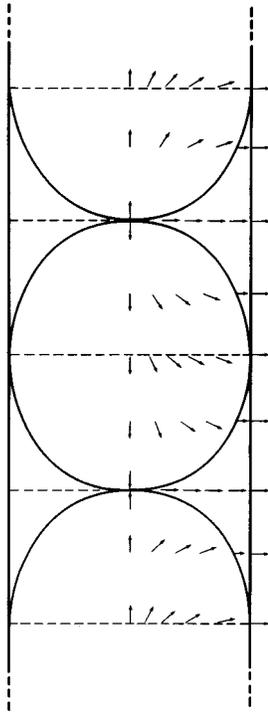


FIG. 4. A topological dipole together with the joints through which the defects are accommodated in a cylinder of height $H=4\sqrt{\alpha\pi}R$. The distance between the defects is $d_c=2\sqrt{\alpha\pi}R$, so that they just *see* each other.

below and above them, two escaped director fields are displayed, pointing in opposite directions.

IV. TOPOLOGICAL DIPOLE

We say that two defects of charges $+1$ and -1 constitute a *topological dipole*. Here we study how they interact while sliding along the axis of a capillary tube. As above, we restrict our attention to a cylinder of height $H=4\sqrt{\alpha\pi}R$, where two defects can be tightly accommodated along with their joints (see Fig. 4). Within our model, there is no point in considering higher cylinders when we deal with a dipole, as the structures here associated with both defects would not interfere with one another as long as the distance d between the defects remains bigger than $2\sqrt{\alpha\pi}R$. To see this better, one should consider that when $d>d_c$, bringing the defects closer to one another would require rearranging the directors so that strips of escaped field between the defects are taken away from them to reappear below the lower joint or above the upper one, while taking the defects farther apart would require bringing back the escaped strips. Clearly, no change in the total elastic energy is involved in either of these rearrangements, and so the defects feel no force; they may freely fluctuate in the capillary, each unaware of the presence of the other. In Fig. 4, $d=d_c$; the defects just *see* each other and are to interact in a way we now describe.

If $d<d_c$, each of the symmetric joints connecting the defects is high $d/2<\sqrt{\alpha\pi}R$. When at equilibrium, they both meet smoothly on a circular cross section of radius

$$\rho R = \frac{d}{2\sqrt{\alpha\pi}}. \quad (13)$$

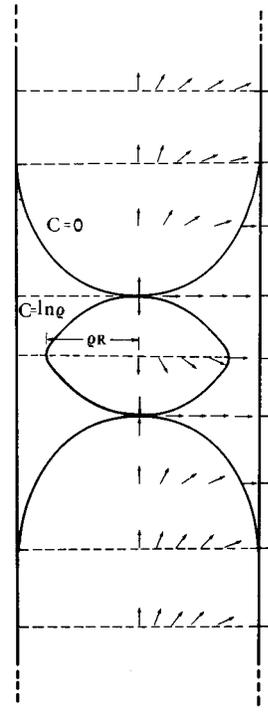


FIG. 5. The equilibrium configuration of the joints describing two defects at a distance $d<d_c$ inside a cylinder of height $H=4\sqrt{\alpha\pi}R$. For the outer joints $\rho=1$, while for the inner ones $\rho<1$.

Figure 5 illustrates one of these equilibrium configurations in a cylinder of height H ; both outer and inner joints are described by a function as in (10) or its opposite, where ρ is set equal to 1 for the outer joints ($C=0$), and is given by (13) for the inner ones ($C<0$). To compute the total elastic free energy $\mathcal{F}(d)$ stored in the cylinder where the defects interact, we apply (12) to the four joints here involved in our construction, and we recall that the energy associated with Cladis and Kléman's director field in the upper and lower strips, each high $(1/2)(H-d-2\sqrt{\alpha\pi}R)$, amounts to

$$2\pi K(H-d-2\sqrt{\alpha\pi}R), \quad (14)$$

where H and R are again the height and radius of the cylinder. Bringing together all these contributions to the elastic free energy $\mathcal{F}(d)$, we obtain

$$\mathcal{F}(d) = 2\pi K\{(3-\ln\rho)\rho\sqrt{\alpha\pi}R - d + \sqrt{\alpha\pi}R + H\}, \quad (15)$$

where ρ depends on d through (13). The derivative of $-\mathcal{F}(d)$ with respect to d is the force felt by each defect

$$f(d) := \pi K \ln \frac{d}{d_c}, \quad (16)$$

which is attractive, being $d<d_c$.

The sign of $f(d)$ has a direct consequence on the absolute energy minimizers; were d free to decrease from d_c to 0, the elastic free energy would decrease as well, indicating that the absolute minimizer would bear no defect, as the defects in a

dipole would annihilate each other for $d=0$. This, however, is a process which needs to be phrased in an appropriate dynamical context.

V. DYNAMICS

Our dynamical model rests upon the dissipation principle on which Leslie [9] has recently rebuilt the classical theory for nematic flows. In the case of interest to us the hydrodynamic macroscopic flow is negligible, as the motion of defects is primarily due to a rearrangement in time of the director field. Thus Leslie's dissipation principle takes a simpler form,

$$\dot{\mathcal{F}} + \mathcal{W} = 0, \quad (17)$$

saying that the time rate of the elastic free energy \mathcal{F} stored within a fixed region in space balances out the energy \mathcal{W} dissipated in the same region by the viscous torques acting on the director. As is customary in nematic liquid crystals, the molecular inertia being negligible, no account is taken in (17) for the kinetic energy of the director motion. Let W be the energy dissipated per unit volume. In the absence of flow, for a director field represented as in (2), W reduces to

$$W = \gamma_1 \left(\frac{\partial \varphi}{\partial t} \right)^2, \quad (18)$$

where $\gamma_1 = \alpha_3 - \alpha_2 > 0$ is the difference between two Leslie coefficients (cf., e.g., Chap. 5 of [12]).

Another assumption is essential to our development. We take each of the configurations traversed by a topological dipole while the defects come closer to each other to be the equilibrium configuration corresponding to the current distance between the defects. Thus (17) will eventually turn into a first-order differential equation for d .

To compute the energy dissipated in the same cylinder of height H where in the preceding section we found the equilibrium configurations for the director, we first need to consider a generic joint as in Sec. II, but movable, so that the angle φ may depend on time through the joint itself. This is achieved by letting r_0 in (3) depend also on t , besides z . Precisely as we arrived at (4), we can now compute the energy \mathcal{W}_h dissipated in a joint of current height h ; it is given by

$$\mathcal{W}_h = 2\pi\gamma_1\alpha \int_0^h \left(\frac{\partial r_0}{\partial t} \right)^2 dz, \quad (19)$$

where α is the same magic number as above. Like (12), also this integral can more conveniently be expressed in terms of z_0 , the inverse function of r_0 defined in (10), which now depends on t too, as does ρ

$$\mathcal{W}_h = 2\pi\gamma_1\alpha \int_0^{\rho R} \frac{\left(\frac{\partial z_0}{\partial t} \right)^2}{\left| \frac{\partial z_0}{\partial r} \right|} dr. \quad (20)$$

We compute an integral like this for each joint in Fig. 5, taking care, in representing the various z_0 's, to regard as

immobile only the center of symmetry of the dipole and to take ρ as 1 or as in (13), according to which joint is being considered. Adding them up, for the total energy dissipated in the process we obtain

$$\mathcal{W} = 2\pi\gamma_1(\alpha\pi)^{3/2}R^3(\lambda\rho+1)\dot{\rho}^2, \quad (21)$$

where λ is defined by

$$\lambda := \frac{4}{\pi^{3/2}} \int_0^{+\infty} \{2e^{-3x^2} + \pi\phi^2(x)x^2e^{-x^2}\} dx \quad (22)$$

in terms of the probability integral

$$\phi(x) := \frac{1}{\sqrt{\pi}} \int_0^{x^2} \frac{e^{-y}}{\sqrt{y}} dy. \quad (23)$$

A numerical computation yields $\lambda \approx 1.445$.

In (21) ρ actually depends on d through (13), and so it decreases as d does. Thus the *effective viscosity* $\gamma := \gamma_1(\lambda\rho+1)$ decreases as the defects approach one another, suggesting that their motion should become faster and faster as time elapses.

Since here $\dot{\mathcal{F}} = -f(d)\dot{d}$, combining (17), (16), and (21), we deduce the differential equation that within our model governs the dynamics of a topological dipole. It is convenient to express it as an equation for the dimensionless parameter ρ , rather than for d

$$T(\lambda\rho+1)\dot{\rho} = \ln\rho, \quad (24)$$

where

$$T := \alpha\pi \frac{\gamma_1 R^2}{K}, \quad (25)$$

is a *relaxation time*, which depends on both the material and the size of the capillary. Collecting data from pp. 105 and 231 of [12], we easily see that for *p*-methoxybenzylidene-*p*-*n*-butylaniline (MBBA) at room temperature in a tube of radius approximately 100 microns, T is of the order of minutes.

VI. ANNIHILATION

We now describe the evolution in time of the distance between the defects. Let $d_0 < d_c$ be the initial value of d ; it corresponds to the initial value $\rho_0 = (d_0/2\sqrt{\alpha\pi R})$ of the parameter ρ . An integration by separation of variables in (24) shows that ρ vanishes in a finite time

$$t_a := T \int_{-\ln\rho_0}^{+\infty} \frac{1}{x} (\lambda e^{-2x} + e^{-x}) dx, \quad (26)$$

which is the *annihilation time*. Let

$$\tau := \frac{t_a - t}{T} \quad (27)$$

be the dimensionless *time before annihilation* and $\delta := d/2R$ the distance between the defects measured in diameters. It follows from (24) and (13) that the function expressing δ in terms of τ can be given the parametric form

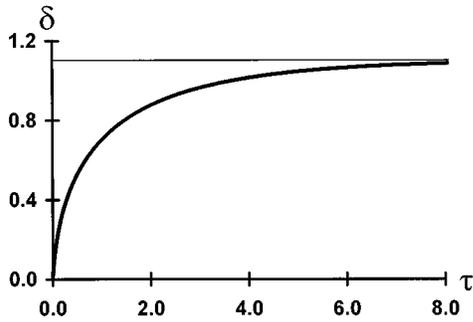


FIG. 6. Graph of the distance δ between the defects measured in diameters versus the dimensionless time τ before annihilation: δ approaches $\sqrt{\alpha\pi}$ as τ diverges to infinity.

$$\delta = \sqrt{\alpha\pi\rho} \quad (28)$$

$$\tau = \int_{-\ln\rho}^{+\infty} \frac{1}{x} (\lambda e^{-2x} + e^{-x}) dx.$$

The graph of δ versus τ is plotted in Fig. 6, as ρ ranges between 0 and 1. It has an asymptote for $\delta = \sqrt{\alpha\pi}$, which it approaches as τ diverges to infinity, whereas it steeply drops into the origin as τ approaches zero.

Further information can be drawn from the dimensionless approaching velocity $v := -\delta T$. By (28) (top line) and (24),

$$v = -\sqrt{\alpha\pi} \frac{\ln\rho}{1+\lambda\rho}, \quad (29)$$

which together with (28) (top line) represent the parametric form of the function plotted in Fig. 7. It diverges logarithmically as the distance approaches zero. The plots in both Figs. 6 and 7 are *universal*, as they do not depend on either the annihilation time or the initial value of d .

After the defects have annihilated each other, the director field in the capillary is still not uniform along the axis, like Cladis and Kléman's. Such a uniform field, which is the absolute energy minimizer, is indeed reached through a further rearrangement that here we describe in words, leaving aside any mathematical detail. The inner joints in Fig. 5 having now disappeared, a possible way to make the escaped

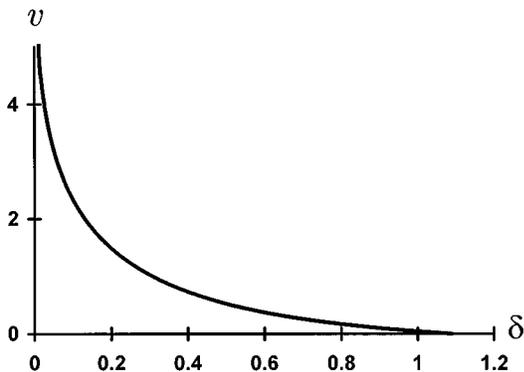


FIG. 7. Graph of the dimensionless approaching velocity v vs the distance δ between the defects measured in diameters: v diverges to infinity as δ approaches 0.

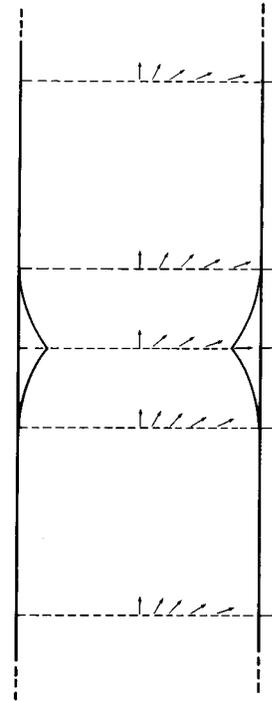


FIG. 8. The rearrangement of the joints after the defects have disappeared. The joints are brought to overlap until a z independent escaped field prevails in the whole region.

field prevail everywhere is by rigidly sliding the outer joints one towards the other, erasing meanwhile the regions where they are thus brought to overlap, as suggested by the sketch in Fig. 8. An easy computation shows that the elastic energy stored in the cylinder steadily decreases in this process.

VII. CONCLUSIONS

We have proposed a mathematical model to describe the approaching and annihilation in a capillary tube of two point defects with topological charges $+1$ and -1 . The most distinctive feature of this model is the interaction force between the defects, which is predicted to depend logarithmically on the distance between them and to vanish when they are farther apart than a critical distance, shortly above a capillary diameter. Such a screening in the interaction might well be an artifact of the special fields employed to describe the director around the defect, though in the absence of any more refined model this issue cannot be unambiguously resolved. We are aware of the other simplifying assumptions upon which our model is built: above all, our treatment of the dynamics, where the motion of the defects is regarded as a sequence of equilibrium states affected by dissipation. We are, however, convinced that the simplicity and predictiveness envisaged here are merits of this model.

In [5] Ericksen reproduces the data of an experiment by Walters and Cladis [13], where the phenomenon we have modeled here was observed for MBBA at 23°C within a capillary tube 150 microns wide. In their experiment the defects move very slowly as long as the distance between them is greater than approximately 1 diameter; then, their relative velocity increases more and more until they annihilate one

another. The data are recorded in a plot which gives distance versus time; they seem to fit nicely the graph in Fig. 6. Nonetheless, we could not go any farther than a qualitative agreement, mainly because of the uncertainty introduced in inferring the data from the experimental plot.

A similar experiment is now being performed in Halle by Hillig and Saupe [14] with a lyotropic liquid crystal. We

hope that their data will soon tell us to what degree the model proposed here can be trusted.

ACKNOWLEDGMENTS

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